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Measurement of mercury mobilization and accumulation in fish in response to prescribed fire in a boreal forest ecosystem

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Introduction

Mercury (Hg) is of great concern in the environment because it biomagnifies up the food chain in aquatic ecosystems (EPA, 2002; EPA, 2000). Mercury is of special concern to residents of Minnesota and the Great Lakes region as evidenced by the advisories on fish consumption issued in Minnesota (MPCA, 2002) and the Lake Superior Binational Program's stated goal of virtual elimination of Hg from the Lake Superior environment. Within Minnesota, the highest Hg levels in fish are found in interior lakes of the northeast including the Superior National Forest and the Boundary Waters Canoe Area (Kaiser et. al. 1996). There are widespread fish consumption advisories in 26 states (including Minnesota), Canada, and Sweden (Glass et al., 1991). Health risks are presumed to be high for people that consume large quantities of fish in these regions. The most susceptible group is prenatal children, followed by young children and women. Numerous animals are also at health risk due to mercury contamination. Hg is implicated in reproductive problems in eagles, otters, mink and other fish-eating animals in the Great Lakes region, and in panther deaths and highly elevated levels of Hg in alligators, bald eagles and raccoons in Florida (Douglas, 1991).

Although we are beginning to understand the cycling of total-Hg and methyl-Hg (bioaccumulative form) in forested watersheds (e.g. Mitchell et al., 2008; Jeremiason et al., 2006; Kolka et al., 2001), little work has been done understanding the role of wildland fire in Hg cycling. A review on Hg cycling did not even address fire as a possible source of atmospherically deposited Hg (Grigal, 2002).

Prior to the work conducted as a result of this study, only two groups of scientists had addressed fire as a possible source of Hg. Hans Friedli and colleagues at the National Center for Atmospheric Research in Boulder, CO assessed the Hg released to the atmosphere following the burning of different types of fuel (Friedli et al., 2001). According to their laboratory studies, nearly 100% of Hg stored in fuels was emitted to the atmosphere with 95% of that emitted as

elemental Hg and particulate Hg accounting for the remainder. Newly released elemental Hg enters the global cycle and undergoes chemical transformations in the atmosphere before being re-deposited. The Hg that we were concerned with in this study was the remaining 5% that is emitted as particulate Hg and has the potential to be deposited locally during a fire event. Subsequent work by this group has assessed actual burns and found upwards of 14% of the mercury released during fires is in the particulate form (Friedli et al., 2003).

A second group of researchers at the University of Montreal had conducted experiments assessing Hg concentrations in the aquatic food chain in undisturbed, burned and logged watersheds in central Quebec (Garcia and Carignan, 2000; Garcia and Carignan, 1999). They found that in lakes where the watershed was burned, there was no difference in zooplankton or northern pike Hg concentrations when compared to lakes in undisturbed watersheds. One drawback of these studies is that data was only collected post-burn and high natural variability in zooplankton and northern pike Hg concentrations could have led to greater uncertainty in their statistics. The mean Hg concentration in northern pike taken from burned lakes (3.0 ug g^{-1}) was considerably higher than those taken from lakes in undisturbed watersheds (1.9 ug g^{-1}), but again, not statistically significant.

Since we were funded a number of other fire-Hg related studies have been published. Six studies have assessed soil and/or vegetation Hg concentrations and emissions resulting from fire. In Florida, annual prescribed fires in longleaf pine systems led to about 0.2 g ha^{-1} yr^{-1} of Hg emitted from soils (DiCosty et al., 2006). In the Sierras, the main source of Hg to the atmosphere following fire was the forest floor with emissions ranging from 0.4 to 5.1 g ha^{-1} (Engle et al., 2006). In the Rocky Mountains of Wyoming, Hg emissions from soils during fires ranged from 3.6-25.3 g ha^{-1} (Biswas et al., 2007). Similar studies in Ontario indicated that plants emitted 98% of the Hg and soils 79% of the Hg they contained, or about 20 g ha^{-1} emitted during fire (Mailman and Bodaly, 2005). Additional studies in Alaska estimated 2 g ha^{-1} of Hg emitted from forest floor only during fire (Harden et al., 2004). Finally, studies in western Canada estimated that increases in peatland fires resulting from climate change will lead to 15X the emission of Hg than current emissions (Turetsky et al., 2006). Other studies have assessed longer range transport of Hg emitted as a result of fire, both from China to the US (Weiss-Penzias et al., 2007) and Canada to the US (Sigler et al., 2003).

Despite some of the more recent research, it is clear from a review of the literature that more investigation needs to be conducted assessing the impact of wildfire or prescribed fire on the Hg cycle and the potential implications on aquatic biota.

This Study

Our study took place in the Boundary Waters Canoe Area Wilderness (BWCAW) in the Superior National Forest (SNF) in the Boreal Region of NE Minnesota. The BWCAW is also a major recreation area in the Upper Midwest, and, because fishing is a preferred activity, Hg levels in fish are of concern. The burning program on the SNF was developed in response to a major blowdown event in July 1999. Fire managers in the Superior Forest developed a fire plan that calls for burning 30,000 of the 400,000 ha of the BWCAW, providing a rare opportunity to study fire/ecosystem Hg interactions. The Boreal Region of NE Minnesota is composed of shallow upland soils and numerous wetlands and lakes, all factors leading to an enhanced potential for Hg transport and bioaccumulation. If a fire effect on Hg cycling and accumulation in fish is to be found, the Boreal Region of Northeastern Minnesota is a location where it could occur. If the null hypothesis is true and fire has no effect on fish Hg concentrations in this

region, we can cross Hg of the list of potential negative impacts of wildfire and prescribed burning programs worldwide. However, if indeed prescribed fire has a deleterious effect on fish Hg concentrations, different fuel reduction strategies, such as mechanized treatment, should be considered. Alternatively, a management program for lake-specific fish consumption advisories could be established following fires.

Project Objective

The primary objective of our research was to determine if fire leads to elevated deposition of Hg and if that deposition increases concentrations in surface waters and ultimately in biota (i.e. fish).

Project Update

We selected undeveloped lakes in watershed that were planned to burned (5 treatment lakes) and not burned (5 control lakes) in and near the BWCAW. We collected precipitation, throughfall, soil and lake water chemistry data (including Hg) and Hg in 1+ year fish from these watersheds both pre-burn and post-burn during 2005-2008. During the timeframe of our funding we anticipated that a minimum of 3 of the 5 watersheds planned to be burned would be lit. Unfortunately during the first 3 years of the experiment, only one watershed was burned. We asked and received a 14 month no cost extension hoping that additional watersheds would burned but again, we were unsuccessful in getting any additional watersheds lit. Our project was dependent upon the Superior National Forest conducting planned prescribed burns in our watersheds. We were unsuccessful in getting additional watersheds burned because in 2006 and 2007 major wildfires occurred in the BWCAW and this consumed the resources necessary to conduct the prescribed burns. In addition there is always the uncertainty of weather conditions, and national budget priorities (e.g. the reallocation of Forest Service resources nationally in 2008 to pay for wildfire suppression costs). Finishing the BWCAW blowdown burns remains a high priority for the Superior National Forest. At this time all four of our watersheds planned to burned are at the top of the priority list of prescribed fires to be conducted.

Although we did not get the fires that we had hoped, we have a data base that is unmatched and we are poised to seek additional funding to continue the study. Because of the delays in the prescribed fires in our watersheds, we have collected multi-year background data in both our treatment and control watersheds, which, when our watersheds are ultimately burned, will give us a better ability to determine the natural variation and should give us more power to determine fire effects. Given the data that we have collected, it would be a travesty if we could not measure following the prescribed fires.

We had hypothesized that fire would increase Hg deposition locally and that deposition would increase lake water concentrations and ultimately fish concentrations. Because we only were able to get one of our treatment watersheds burned we are currently unable to address fire effects questions on lake water and fish. As a result, our current findings address the background data that we collected and the effect of fire on atmospheric deposition and soils.

Important Results

We recently submitted a paper to the journal *Science of the Total Environment* that used our extensive data base on soil and lake chemistry and watershed characteristics to predict fish Hg concentrations among our lakes. Spatial relationships between yellow perch Hg tissue concentration and a total of 45 watershed and water chemistry parameters were evaluated for two

separate years: 2005 and 2006. Results show agreement with other studies where watershed area, lake water pH, nutrient levels (specifically dissolved NO^3-N) and dissolved iron are important factors controlling and/or predicting fish Hg level. Exceeding all was the strong dependence of yellow perch Hg level on soil A-horizon Hg and O-horizon Hg concentrations (Spearman ? = 0.81). Soil B-horizon Hg concentration was significantly correlated (Pearson r = 0.75) with lake water Hg concentration. Lakes surrounded by a greater percentage of shrub wetlands (peatlands) had higher fish tissue Hg levels, thus it is highly possible that these wetlands are main locations for mercury methylation. Stepwise regression was used to develop empirical models for the purpose of predicting the spatial variation in yellow perch Hg over the studied region. The 2005 regression model demonstrates it is possible to obtain good prediction (up to 60% variance description) of resident yellow perch Hg level using upland soil O-horizon Hg as the only independent variable. The 2006 model shows even greater prediction (r^2 = 73%), using lake water dissolved iron and watershed area as the only model independent variables. The developed regression models in this study can help with interpreting Hg concentrations in low trophic level fish species for untested lakes of the greater Superior National Forest and surrounding Boreal ecosystem.

Our data base also includes background throughfall and open precipitation measurements of both total (THg) and methyl Hg (MeHg). We completed a MS student (Emma Witt) and have a paper in press at the journal *Water, Air, and Soil Pollution* that includes this data. Throughfall and open canopy precipitation samples were collected in 2005 and 2006 using passive precipitation collectors from pristine sites located across the SNF. Samples were collected approximately every 2 weeks and analyzed for THg and MeHg. Forest canopy type and density were the primary influences on THg and MeHg deposition (Figures 1-3). Highest THg and MeHg concentrations were measured beneath conifer canopies (THg mean=19.0 ng L^{-1}; MeHg mean=0.28 ng L^{-1}) followed by deciduous throughfall (THg mean=12.5 ng L^{-1}; MeHg mean=0.19 ng L^{-1}) then open precipitation (THg mean=8.2 ng L^{-1}; MeHg mean= 0.12 ng L^{-1}). The greater efficiency of conifers at scavenging THg and MeHg from the atmosphere may increase the risk of mercury related water quality issues in conifer-dominated systems.

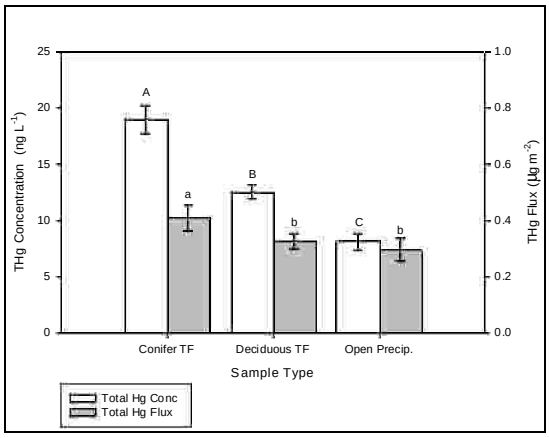

Figure 1. Mean THg concentrations and flux by canopy type. Letters denote significant differences at the 0.05 level; error bars represent one standard error. THg concentrations varied among all three canopy types, while flux was different for conifer throughfall, but not between deciduous throughfall and bulk precipitation.

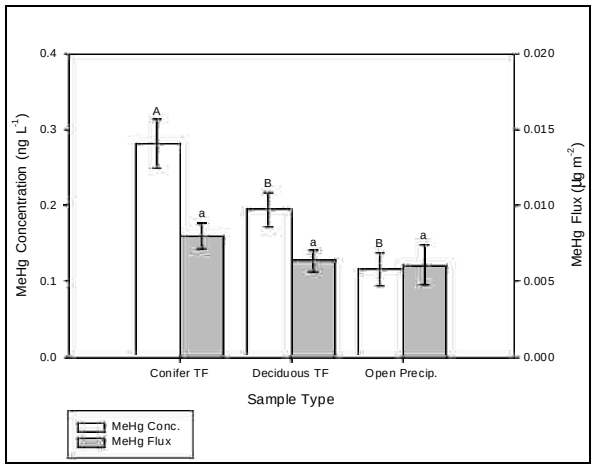

Figure 2. Mean MeHg concentrations and flux by canopy type. Letters denote significant differences at the 0.05 level; error bars represent one standard error. MeHg concentrations differed between conifer throughfall and open bulk precipitation, while MeHg flux was not significantly different among any of the three sample types.

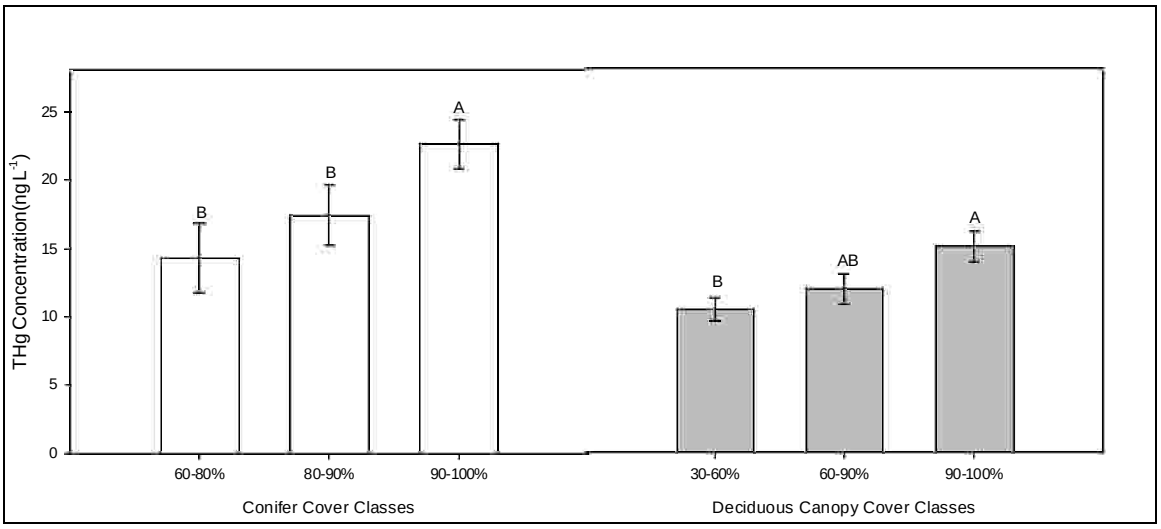

Figure 3. Canopy density effects on THg concentration. Significant differences were found among the different conifer throughfall cover classes ($p = 0.02$, $n = 52$); canopies with densities greater than 90% had significantly higher THg concentrations than canopies with densities less than 80% ($p = 0.008$, $n = 41$) and higher THg concentrations than canopies with densities between 80 and 90% ($p = 0.05$, $n = 39$). THg concentrations from deciduous throughfall were also significantly different based on canopy density ($p = 0.004$, $n = 84$). Samples from canopies with densities greater than 90% had higher THg concentrations than samples from canopies with densities less than 60% density ($p = 0.003$, $n = 63$), but were not different for canopy densities between 60 and 90% ($p = 0.12$, $n = 53$). Error bars represent one standard error.

Because we did have one prescribed fire and several wildfires occur during study, we were able to capture a number of these events with our widely spaced throughfall and open

precipitation collector network. We recently submitted an article to the journal *Environmental Science and Technology* assessing fire effects on atmospheric deposition. We believe this is the first study that was able to collect wet deposition during fire events. We compared background throughfall and open precipitation samples (above data) to those taken when smoke plumes were over our collectors. Like the above paper, throughfall and open precipitation samples were collected pre- and post-fire in 2005 and 2006 using passive precipitation collectors across the SNF. Samples were collected approximately every two weeks and analyzed for total Hg (THg) and methyl Hg (MeHg). THg concentrations increased significantly post-fire in conifer throughfall (>4X increase), open precipitation (2.5X), and when all canopy types were considered (2.9X) (Figure 4). MeHg concentrations also increased significantly after fire regardless of the cover type (conifer throughfall: 10X increase; open precipitation: 3.5X increase; deciduous throughfall: 1.9X increase; all canopy types analyzed together: 8X increase) (Figure 4). THg deposition increased significantly under conifer cover (3.8X). MeHg deposition increased significantly after fire when all canopy types were analyzed together (4.6X) and in conifer throughfall (5.9X). Canopy type influenced the magnitude of post-fire THg and MeHg increase and the duration of elevated MeHg levels. Particulate Hg present in forest fire smoke represents a short-term source of increased Hg in the atmosphere that is available for local re-deposition during and following fire.

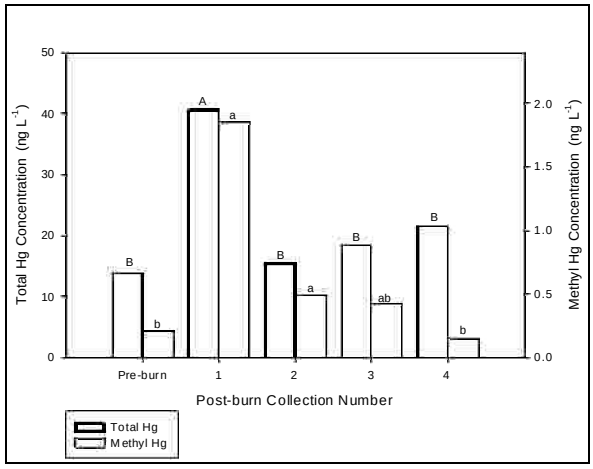

Figure 4. Mean THg and MeHg concentrations for pre-burn and post-burn collections for all sample types. Significant increases were measured in both THg and MeHg concentrations in the first sample following fire; significant increases persisted in MeHg concentrations until after the second post-burn collection.

Drs. Woodruff and Cannon sampled soils in the prescribed burned watershed as well as in some of the other burned watersheds in the SNF and Voyageurs National Park (VNP). They are currently working on a manuscript assessing the effects of wildfire on soil mercury sequestration and mercury cycling across the region. Their work indicates that mercury in forest soils is strongly influenced by forest disturbance, especially forest fires. A study of the Section 33 wildfire, which burned over 400 ha in VNP in 2004 measured mercury in soils across gradients of fire severity. Soil mercury losses in high fire-severity areas, when compared to adjacent unburned areas, were: 1) ~ 0.22 mg Hg m^{-2}, because of complete combustion of an O horizon, and 2) ~ 0.6 to 0.9 mg Hg m^{-2} from combusted A-horizon mineral soils. Samples

collected two weeks after the fire and prior to any significant rainfall, indicated that these Hg losses were an immediate and direct consequence of combustion and volatilization of Hg from the soil. Such Hg losses from soil resulting from severe wildfires are likely to be gradually ameliorated by continued atmospheric deposition and sequestration which eventually restores Hg contents toward pre-fire levels. An outgrowth of this JFSP study in the BWCAW demonstrated that the time over which Hg concentrations in soils can increase after a fire is many decades or even centuries. O-horizon and A-horizon soils from 11 areas of primarily virgin boreal forest within the BWCAW sampled for the JFSP study have fire-origin stand ages (time since last severe wildfire) ranging from the 1700's to the 1970's. Integrated with our VNP data, there is a highly significant relationship between fire-origin stand age and mean Hg concentration in A-horizon soils (r^2 = 0.87) in which older stands have progressively higher Hg concentrations. This relationship suggests that the pattern of Hg concentrations in forest soils should form a mosaic that varies in accordance with the mosaic of wildfire-controlled stand ages. The fire-origin stand age within a watershed thus provides a means of predicting the Hg load in soils, particularly where the stand age mosaic has not been modified by other human activity.

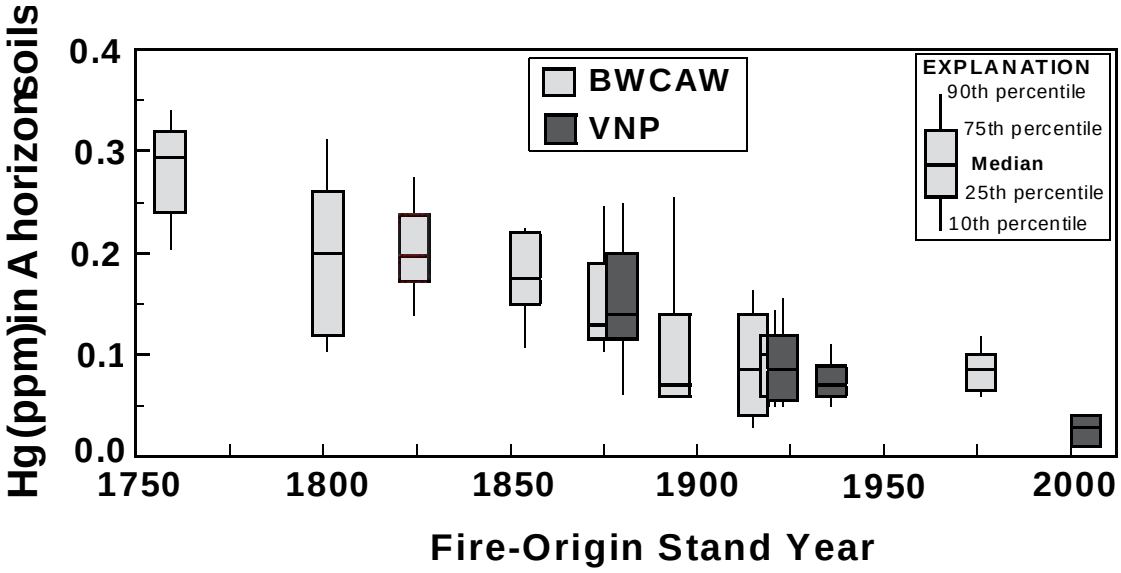

Figure 5. Total mercury concentrations for mercury in A-horizon soils from the Boundary Waters Canoe Wilderness Area (BWCAW) and from Voyageurs National Park (VNP) plotted again fire-origin stand year.

Products

Theses

Emma Witt, M.S., University of Minnesota, Department of Soil, Water and Climate. 2007. Thesis Title: Atmospheric Deposition of Total and Methyl Mercury in Boreal Forests and Response to Forest Fire. 54 pp.

Journal Papers

Witt, E.L., R.K. Kolka, E.A. Nater, and T.R. Wickman. 2008. Influence of the forest canopy on total and methyl mercury deposition in the boreal forest. Water, Air, and Soil Pollution (in press)

Witt, E.L., R.K. Kolka, E.A. Nater, and T.R. Wickman. 2008. Forest fire effects on mercury deposition in the boreal forest. Environmental Science and Technology (submitted)

Gabriel M.C., R. Kolka, T. Wickman, E. Nater, and L. Woodruff. 2009. Evaluating the spatial variation of total mercury concentrations in young-of-year fish for watershed-lake systems within the southern Boreal Shield. Science of the Total Environment (submitted)

Presentations

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Conclusions

Although we planned to have more prescribed fires in our treatment watersheds to test our hypotheses related to fire effects on lake water and fish concentrations, we have built an impressive data base on background concentrations of Hg in atmospheric deposition, soils, lake water and fish. In addition, we have shown for the first time in the literature a fire effect on local atmospheric deposition. Based on our data, we estimate that in the year of a fire a one-time pulse of mercury is deposited that is equal to an additional 30-40% of the amount normally deposited. Our future studies will enable us to tell if this additional Hg is enough to change concentrations in lake water and fish.

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